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Title: Studies of Fission-Induced Surface Damage in Actinides Using Ultracold Neutrons

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Studies of Fission-Induced Surface Damage in Actinides Using Ultracold Neutrons

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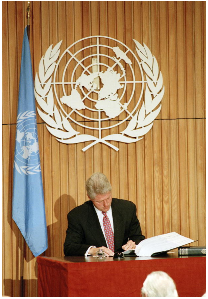
March 6, 2014

Nuclear Safety in the Modern Era

Comprehensive Nuclear Test-Ban Treaty

- United States signed on September 24, 1996
- Ratification dependent on **Science Based Stockpile Stewardship Program** to ensure safety and reliability of nuclear weapons in active stockpile



Stockpile stewardship

- No full-scale nuclear weapons testing since 1992
- How does aging of nuclear material affect reliability of nuclear weapons?
- NNSA mission: *Maintain the safety, security, and effectiveness of the nuclear deterrent without nuclear testing*

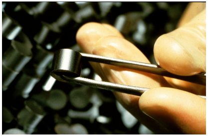
Major Effort at LANL

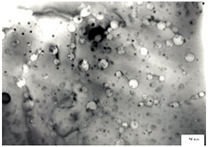
- DARHT: X-ray radiography for non-nuclear tests
- LANSCE: spallation neutrons
- pRAD: proton radiography
- TA-55

Aging of Nuclear Material

Nuclear energy

- Stopping of fission fragments in UO_2 = primary source of heating
- Irradiations causes "fission spikes" $\rightarrow$ atomic displacements
- Physical and chemical properties of fuel change over time: reduced thermal conductivity, enhanced diffusion, creep, and gas release
- Formation/resolution of fission gas bubbles[a]
- UO_2 pellets in fuel rods: porosity gradually destroyed by fission tracks





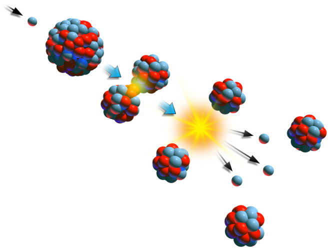
[a] J. App Phys **44** 3575 (1973)

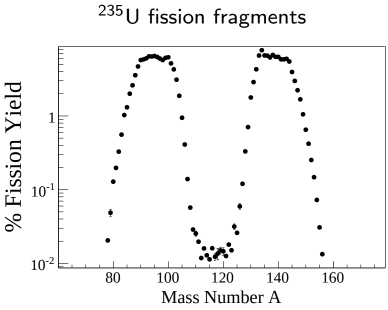
Damage from Material Sputtering

- Insulators vs. conducting material
- Semiconductors and metals in microelectronic components
- Space science: Predicting lifetime of unshielded detectors in space

Fission Process

Fission Studied Extensively, Well Understood





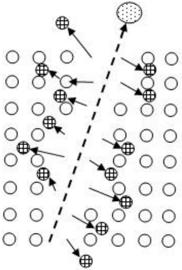
Typically 2 fragments emitted

- E$\sim$100 MeV, $\frac{v}{c} \sim 10\%$
- A$\sim$100
- range$\sim$10μm

After-effects of fission?

Fission fragments

- Very heavy, very energetic, charged particles
- How is energy deposited in material?
- Damage to the material?
- Near material surface: ejection of matter



Macroscopic description

- $>$ 90% energy transferred to electrons
- Rapid ionization $\rightarrow$ rapid recombination
- Intense local heating
- Nuclear collisions = atomic displacements (damage)

Energy Transfer of Fission Fragments

Microscopic description

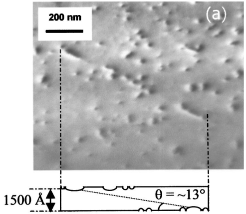
- No complete quantum-mechanical model
- Binary Collision Approximation: simple description of inelastic scattering
- Several "Thermal Spike" models: abrupt rise in temperature in cylindrical region around track
- "Coulomb Explosion" model: high charge density → expansion
- Speculation: formation of shock wave?

Differentiating between models

- CEM predicts more point defects per ion, higher temperatures of ions near particle track
- Yield and angular distribution of sputtered atoms
- Depth effects: range of fission fragments → sputtering

Fission tracks

- Irradiated in reactor by thermal neutron flux
- Fission fragment near surface → visible deformation
- Attempts to characterize fission tracks in UO_2 pellets and thin films[a]



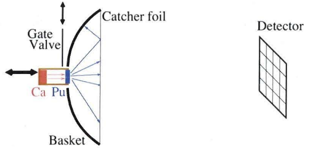
[a] J. App Phys **44** 3575 (1973), J. App Phys **92** 5837 (2002)

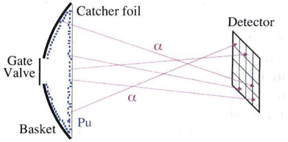
Sputtering

- Ejection of material from surface
- Many previous measurements of sputtered atoms per fission
- **Significant disagreement in yield, distribution!**

Recent Efforts at LANL

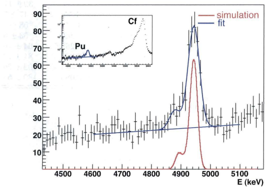
Sputtering Yield of Pu bombarded by ^{252}Cf Fission Fragments

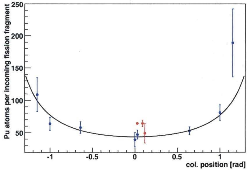




Results

- Fission fragments from ^{252}Cf enter back side of Pu sample
- 32 μm thick Pu foil
- Measured 63±1 sputtered atoms/fission
- BCA predicts no sputtering through thick Pu foil; forward-directed angular distribution





Inducing Fission with Ultracold Neutrons

New Technique for understanding sputtering

- Induce fission using Ultracold Neutrons
- Excellent control of neutron energy
- Very sensitive probe of fission as function of depth

LANL: Unique Position for work

- LANSCE: one of world's brightest sources of UCN
- Expertise in fabrication and analysis of actinide targets

Ultracold Neutrons

Class	Energy	Source
Fast	> 1 **MeV**	**Fission reactions / Spallation**
Slow	**eV – keV**	**Moderation**
Thermal	**0.025 ev**	**Thermal equilibrium**
Cold	μ**eV – meV**	**Cold moderation**
Ultracold	$\leq$ **300 neV**	**Downscattering**

How cold is Ultracold?

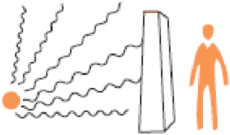
- Temperature < 4 mK
- Velocity < 8 m/s
- Usain Bolt ~ 12 m/s

UCN can be bottled

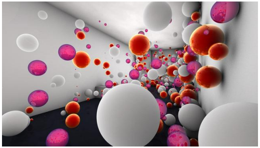
- Gravitational ($V = mgh$): 100 neV / meter
- Magnetic ($V = -\vec{\mu} \cdot \vec{B}$): 60 neV / Tesla
- Material $\left(V = \frac{2\pi\hbar^2 Nb}{m}\right) \begin{cases} {}^{58}\text{Ni}: & 335\text{ neV} \\ \text{DLC}: & 250\text{ neV} \\ \text{BeO}: & 250\text{ neV} \\ \text{Cu}: & 170\text{ neV} \end{cases}$

Why are Ultracold Neutrons special?

They bounce!



Typical radiation



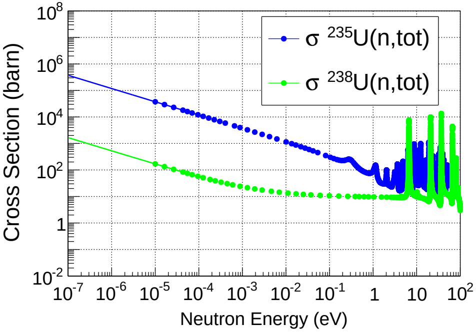
Ultracold Neutrons

Features of UCN experiments

- Usually fundamental physics studies, not yet material science studies!
- Defined decay volume
- 100% polarized using magnetic fields; limited depolarization due to material surfaces
- Low neutron-generated backgrounds
- Pulsed beam: limits backgrounds
- However, limited statistics

UCN-Induced Fission

Uncharted energy regime (10^{-7} eV)



Very high theoretical cross section: $\sigma \sim \frac{1}{v}$

Cross Section (barns)			
UCN Energy	200 neV	300 neV	400 neV
^{235}U(n,tot)	2.64×10^5	2.16×10^5	1.87×10^5
^{238}U(n,tot)	1.17×10^3	9.57×10^2	8.29×10^2

UCN Penetration Depth

Material potential

- UCN are reflected by optical potential of material
- Fermi potential $V_F = \frac{2\pi\hbar^2}{m_n} Nb$

	V_F (neV)
^{235}U	135
^{238}U	110
UO_2	125
UO_3	75–120
U_2O_8	110

Material Depth

UCN range in foil (μm)				
Comp.	% ^{235}U	200 neV	300 neV	400 neV)
DU	0.2%	118	144	191
NatU	0.7%	66	81	101
SEU	2%	31	38	45
LEU	5%	13	17	20
HEU	20%	4	4.5	5
	100%	0.8	0.9	1

Program Goals

Objective: Differentiate between models

Characterize sputtered material...

- Yield per fission
- Mass distribution
- Energy distribution
- Angular distribution
- Charge composition?

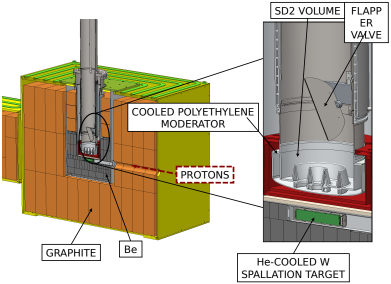
as a function of...

- UCN-energy
- Material thickness
- Material surface: effect of oxide layer
- Alloys, material layers
- Expand to more actinides

UCN Production at LANSCE

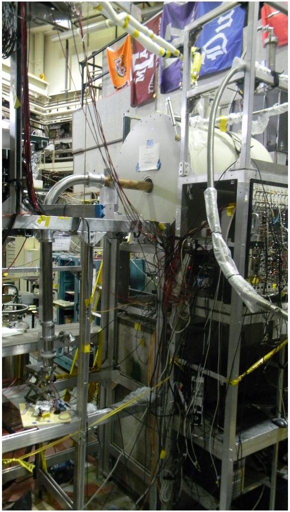
The LANSCE UCN Source[1]

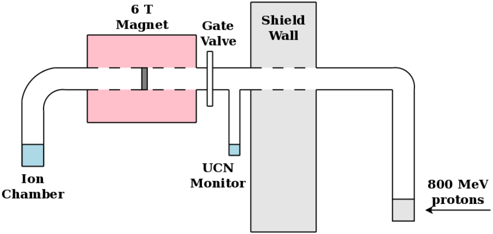
- 800 MeV proton beam + tungsten target → spallation neutrons
- Single scatter in solid deuterium: CN→UCN + phonon
- Remove phonons: SD_2 cooled to 4K
- "Flapper" valve shields UCN from SD_2
- High density at shield wall: 50 UCN/cc
- Pulsed beam: Low background



[1] Rev. Sci. Instrum. 84, 013304 (2013)

Experimental Area





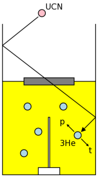
Detection

- UCN Monitor = Normalize for fluctuations in UCN production
- Gate valve permits UCN entry to experiment
- 6 T magnet = near 100% polarization
- UCN drop through Al window into ion chamber

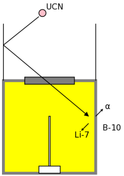
Ion chamber

Several modes of operation

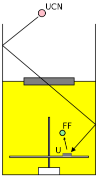
- Mode 1: Counting ultracold neutrons
  - $^{3}\mathrm{He} + \mathrm{n} \rightarrow \mathrm{p} + \mathrm{t}$
  - $^{10}\mathrm{B} + \mathrm{n} \rightarrow \alpha + {}^{7}\mathrm{Li}$
- Mode 2: Counting fission fragments
- Mode 3: Exposure (sputtering)



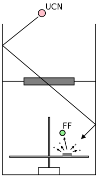
Mode 1a



Mode 1b



Mode 2



Mode 3

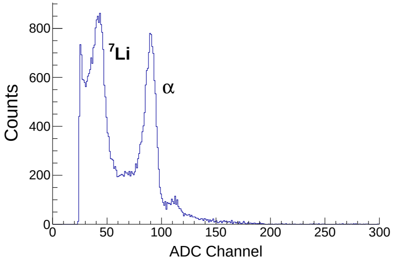
Counting UCN

UCN Beam Monitor[2]

- ^{3}He filled multi-wire proportional chamber
- 50% transmission through window into detector; 80% efficient

Baseline UCN Rates[3]

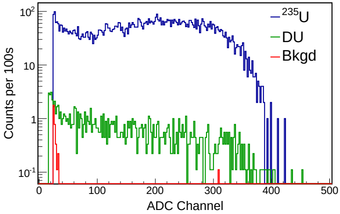
- Boron-coated cylindrical ion chamber, 1 barr argon
- Near 100% efficient for UCN entering chamber
- Rate: 4.5kHz (for 125 Hz beam monitor rate)



[2] Nucl. Instrum. Meth. Phys. Res. A **599** 248 (2009)
[3] Nucl. Instrum. Meth. Phys. Res. A **691** 109 (2012)

Experiment

- Identical experimental setup
- Cylindrical ion chamber with boron coating removed
- Effect of UCN bottling?
- 200 mbarr argon: α's range out



^{238}U

- 2.25 cm diameter, 1 mm thick disk of Depleted Uranium ($\sim 0.2\%$ ^{235}U)
- Rate: $(1.3 \pm 0.8) \times 10^{-4}$ fission/UCN

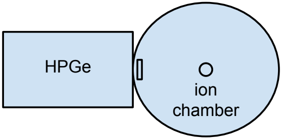
^{235}U

- 2.2 cm diameter, 1 mm thick disk of HEU ($> 80\%$ ^{235}U)
- Rate: $(1.90 \pm 0.02) \times 10^{-2}$ fission/UCN

Neutron Capture Gammas

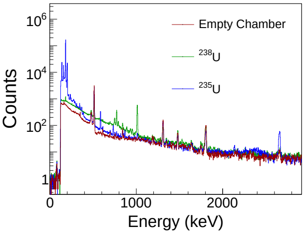
HPGe detector

- Calibration: ^{60}Co and ^{137}Cs gamma sources
- Goal: tag gamma, look for fission



Observed Spectra

- Empty chamber with/without UCN: additional 480 keV line from residual Boron coating
- Decay gammas from ^{235}U/^{238}U observed; some additional lines
- No additional gamma lines with UCN?



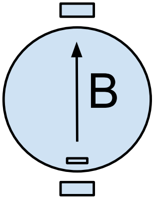
Neutron Spin Dependence

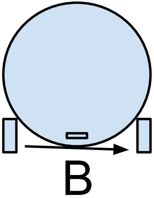
Neutron Polarization

- 6 T Magnet: near 100% UCN polarization
- Neutron spin aligned with field

Experiment

- Neodymium magnets installed on chamber: $\vec{B}$ field normal and parallel to surface
- $\sim$200G field normal to surface: $(1.92 \pm 0.02) \times 10^{-2}$ fission/UCN
- $\sim$50G field parallel to surface: $(1.94 \pm 0.02) \times 10^{-2}$ fission/UCN
- No magnets: $(1.90 \pm 0.02) \times 10^{-2}$ fission/UCN

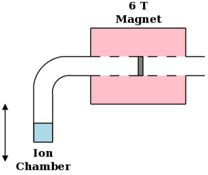




Adjusting UCN Energy with Gravity

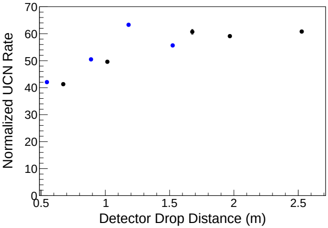
Two primary methods for control

- PPM: Magnetic field (60 neV/T)
- Gravity (100 neV/m)



Effect on UCN Rate

- Adjust height of ion chamber
- Sensitive to geometry (type of elbow)
- Sensitive to UCN guide quality
- Al window: 50 neV loss

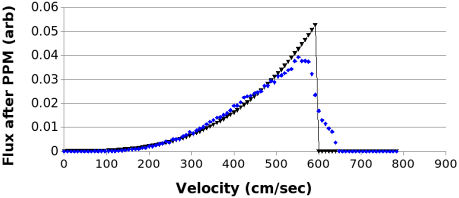


Simulating UCN Monte-Carlo

UCN Monte-Carlo

- Assume UCN energy distribution $v^2 dv$
- LANSCE beam structure
- Propagate UCN through guide system
- Empirical loss per bounce, specularity

UCN energy spectrum after PPM

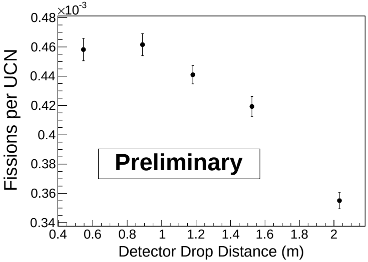


UCN-energy dependence of fission rates

Depleted uranium sample

- Electropolished (thin oxide layer) 2.25 cm diameter, 1 mm thick disk sample[4]
- Identical geometry to ^{3}He gravity scan
- Use beam monitor normalization

Measured fission rate decreases as UCN energy increases

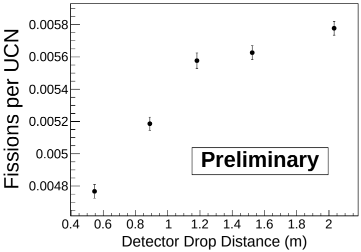


[4] Thanks to J. Cooley, MST

^{235}U sample

- Thin sample: 30 μg heavily oxidized ^{235}U on tape
- Identical geometry and normalization to ^{3}He gravity scan

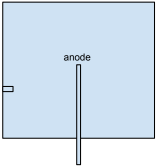
Measured fission rate increases as UCN energy increases

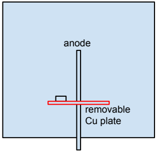


Evidence of Sputtering

Evidence of UCN-induced sputtering?

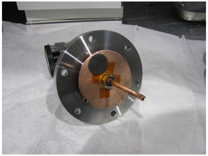
- Installed 2.2 cm diameter, 1 mm thick disk of heavily oxidized ^{235}U for $\sim$20 minutes
- Exposed to UCN for $\sim$10 minutes
- Removed sample: small signal still observed!
- α rate = 2.63$\pm$0.07 Hz ($\sim 10^{17}$ atoms)





Check: No UCN exposure

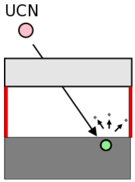
- ^{235}U installed on removable copper plate: reduce chance of contamination
- Installed for $\sim$15 minutes, not exposed to UCN
- Removed copper plate with sample
- α rate = 0.78$\pm$0.04 Hz ($\sim 10^{16}$ atoms)
- Inconclusive: contamination? α-induced sputtering? chamber pumping/pressurizing?



UCN Exposure Studies

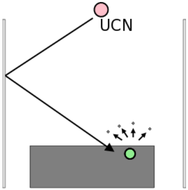
Exposure to Silicon wafers

- 1" diameter, 475 μm thick, polished wafer
- Installed in evacuated ion chamber, exposed to UCN
- Exposed to 2.2 cm diameter, 1 mm thick electropolished DU disk



Exposure to Ni cylindrical foil

- 0.005" Ni foil, 1.15" diameter cylinder, 2.835" height
- UCN bottle: Ni material potential $\sim$ 300 neV
- Exposed to DU disk and 30 μg ^{235}U thin sample

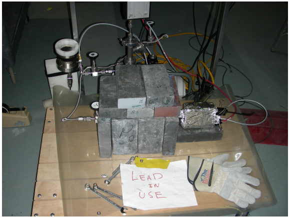


Sputtering on Silicon

Experiment A

- DU + Si assembly total exposure time: 37.5 hours
- 3×10^7 total UCN in chamber
- α rate on wafer: 0.0022 ± 0.0003 Hz $\rightarrow$ 0.18 μg ^{238}U

Counting Chamber



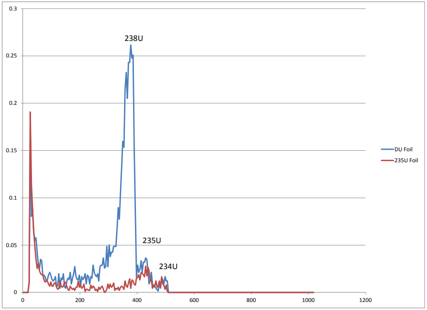
Sputtering on Nickel

DU-exposed Ni foil

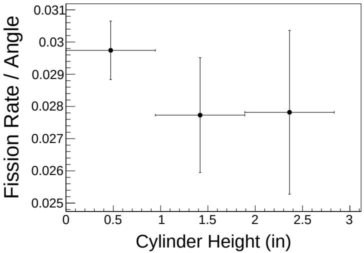
- Total exposure time: 9.5 hours
- 10^8 integrated UCN in chamber
- α rate on foil: 0.040 ± 0.001 Hz $\rightarrow 3\ \mu$g ^{238}U

^{235}U-exposed Ni foil

- Total exposure time: 18 hours
- 10^8 integrated UCN in chamber
- α rate on foil: 0.0058 ± 0.0003 Hz $\rightarrow 0.07\ \mu$g ^{235}U



Sputtering on Nickel: Angular Distributions



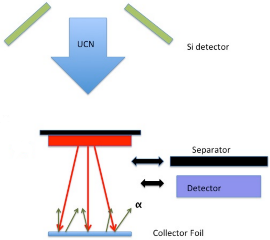
DU-exposed Ni foil

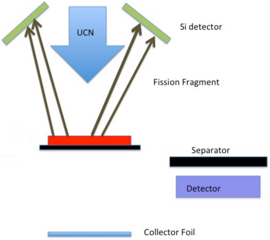
- Sputtering distribution $\sim$ isotropic

Next:

- Self-sputtering? Expose Si wafers, Ni foils *without* UCN
- Observe ejected material with AFM/TEM?

Characterize Ejected Material In-Situ





Important questions:

- How much comes off?
- Size distribution vs. depth/surface quality?
- Kinetics vs. depth?

Summary

First observation of UCN-induced fission

- Previously no fission data at these energies
- Initial characterizations of UCN energy dependence, material thickness

First observation of sputtering from UCN-induced fission

- Proof of principle demonstrated
- Initial characterizations of sputtered rates, angular and size distribution underway

Future plans:

- Determine absolute cross sections
- Develop scheme for characterizing ejected material
- Explore effect of material thickness, surface preparation in more detail
- Develop path forward for plutonium and other actinides